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DEVELOPMENT OF IMPROVED HEAT STERILIZABLE POTTING COMPOUNDS

Third Quarterly Report Covering Period
1 January 1964 to 31 March 1964

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DEVELOPMENT OF IMPROVED HEAT STERILIZABLE POTTING COMPOUNDS

[redacted]

by

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George C. Marshall Space Flight Center
NASA, Huntsville, Alabama

Approved: ______________________
W.H. Colner, Manager
Materials Technology Department

AEROSPACE GROUP
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[redacted]

FOREWORD

This report summarizes the work performed under the National Aeronautics and Space Administration's Contract NAS 8-5499, "Research and Development of Improved Heat Sterilizable Potting Compound," for the period 1 January 1964 to 31 March 1964. This work is sponsored by the Engineering Materials Branch of the Propulsion and Vehicle Engineering Division, George C. Marshall Space Flight Center, Huntsville, Alabama. Mr. John T. Schell is the NASA Project Engineer on this program.

ILLUSTRATIONS

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ABSTRACT

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A vacuum train is being fabricated in which the materials which passed the preliminary screening test will be further evaluated. The train is pumped in three stages by a mechanical roughing pump, by a mercury diffusion pump, and cryogenically by liquid nitrogen. The specimens exposed to the low pressure environment created by this train will be measured for dielectric constant, dissipation factor, and insulation resistance at regular intervals over a 500 hour test span. The test specimens will be maintained at 150°C.

Infrared spectra of the prepolymers and their crosslinking agents have been taken from the materials which will be exposed to the low pressure thermal environment of the test vacuum train. Provisions have been made to trap and analyze by infrared spectroscopy any condensable gases or liquids emanating from the test specimens. The spectra of these materials will then be compared to the spectra of the starting materials, to determine the source of the condensible materials.

Samples of quartz have been received from various vendors for evaluation as fillers for the best resins. A small synthesis effort has been designed to make these fillers more compatible with the resins through the development of suitable finishes.

The molecular still has been set up. This device will be available for the modification of any suitable materials from the preliminary evaluation test.

Preliminary screening tests have been completed on Shell Epon H25 and Epon X24. At the suggestion of project management from the Marshall Space Flight Center, several alternate cures for Dow Corning Sylgard 182 were tried. The data indicated that the suggested cures produced materials which were equivalent to those produced by the cures used by the Hughes Aircraft Company earlier in the program.

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During this report period the work on thermal treatment for moisture absorption recovery was completed.

An important discovery bearing very heavily on the goals of this program has been made in the course of an investigation into materials for use in the Phoenix missile and the AMCS for the F-111A. The discovery is a mathematical equation which accurately predicts the thermal coefficient of expansion of a composite system from the coefficients of thermal expansion of the resin and the filler. The measurements which correlate with the calculated values were all made at temperatures below the Tg of the investigated materials. The use of this equation aids in the formulation of the low coefficient of thermal expansion materials required by this program and should prove most valuable in saving investigation time.

author

PROGRAM PLAN AND ACCOMPLISHMENTS

The Hughes approach to NAS 8-5499, "Research and Development of Improved Heat Sterilizable Potting Compound," is divided into six phases. The literature search is complete.(1) The preliminary screening phase is complete, and all the candidate materials have been selected for submission to a simulated space environment. The most promising materials will be modified according to the information found in the literature search and gleened from the experimental procedures. The modified materials will be submitted to the tests specified in the request for proposal.(2) Recommendations for future fruitful work will be made on the basis of the results of this program.

PRELIMINARY SCREENING

The preliminary screening test was divided into two parts. The results of both tests were pooled to determine the best candidate materials for further evaluation. These tests are described in the Second Quarterly Report.

EXPERIMENTAL PROCEDURES FOR PRELIMINARY EVALUATION TESTS

Vacuum trains were fabricated for the Preliminary Evaluation Test phase of the program. The trains consist of a double manifold, which is consecutively pumped in three stages, a mechanical pump, a mercury diffusion pump, and cryogenically pumped stage using liquid nitrogen. The trains have a capacity of 25 specimens. The test specimens are four rod embedded electrode dielectric test specimens.(3, 4, 5, 6) The specimens are prepared, measured in the ordinary measuring fixture, then shielded and assembled into the glass test chambers with coaxial leads and thermocouples. The glass test chambers are then assembled to the vacuum trains with heaters to provide the appropriate thermal environment for the test. The details of the assembly of the constituent parts of the vacuum train test cells are shown in Figures 1 through 5.

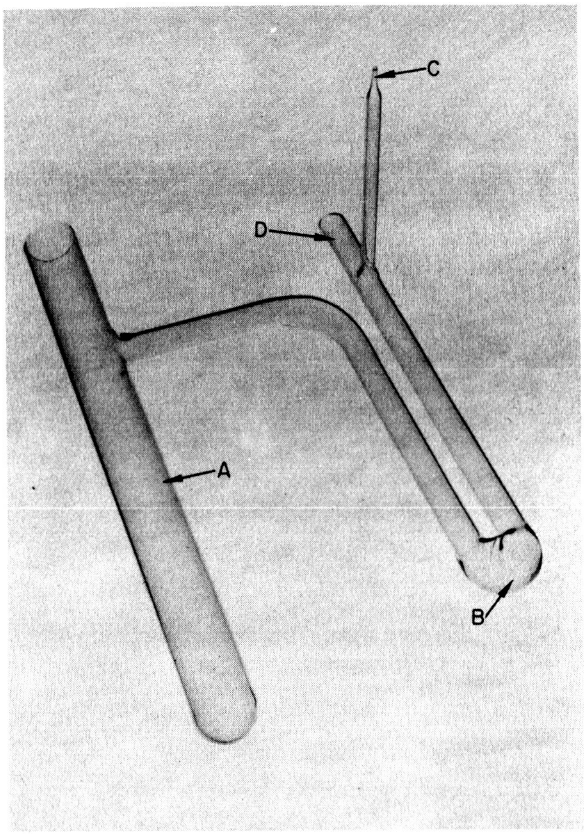


Figure 1. Four rod, embedded electrode test chamber. A - specimen chamber; B - U-shaped liquid nitrogen cryotrap; C - sealed tip used to extract gaseous samples for spectroscopic analysis after test; D - to exhaust manifold.

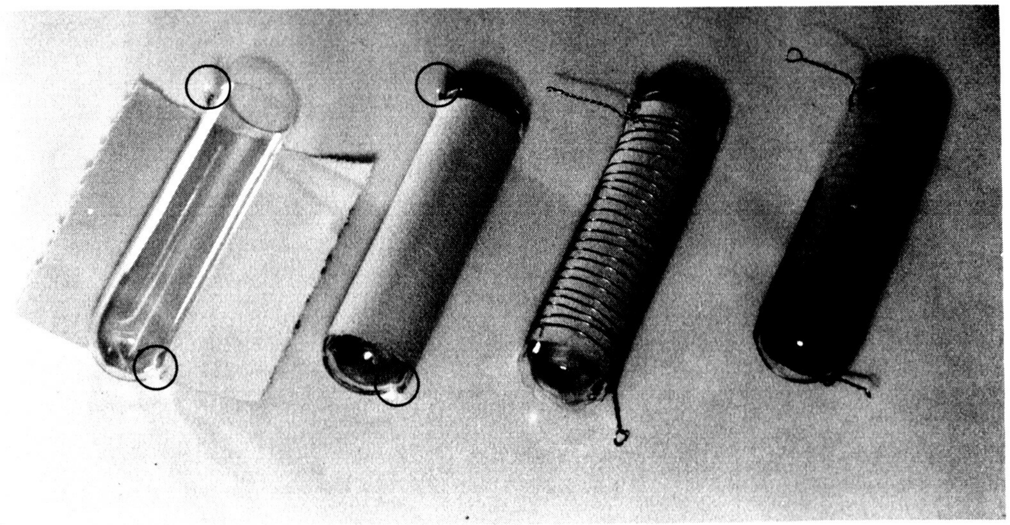


Figure 2. Construction of heating ovens. Resistance wire is wrapped around a pyrex thimble to form an oven for the thermal exposure of the preliminary evaluation tests. Left to right: thimble and asbestos paper; thimble wrapped with asbestos paper (note circled hook-shaped anchors for the nichrome wire); thimble wound with nichrome wire; completed oven.

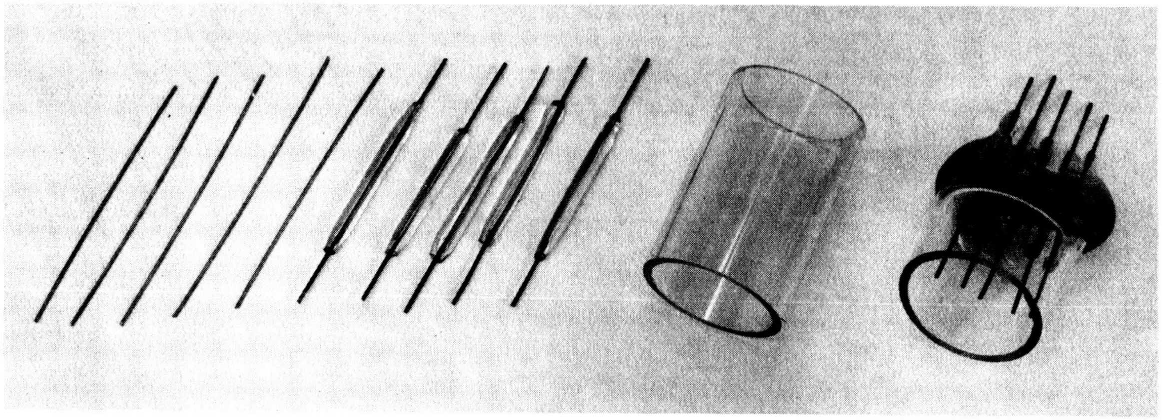


Figure 3. Fabrication of uranium glass-tungsten graded glass seals. Left to right: tungsten pins; tungsten pins with vacuum drawn uranium glass sheaths; uranium glass tube; tungsten conductor pins for embedded electrode and thermocouples sealed in test chamber cap.

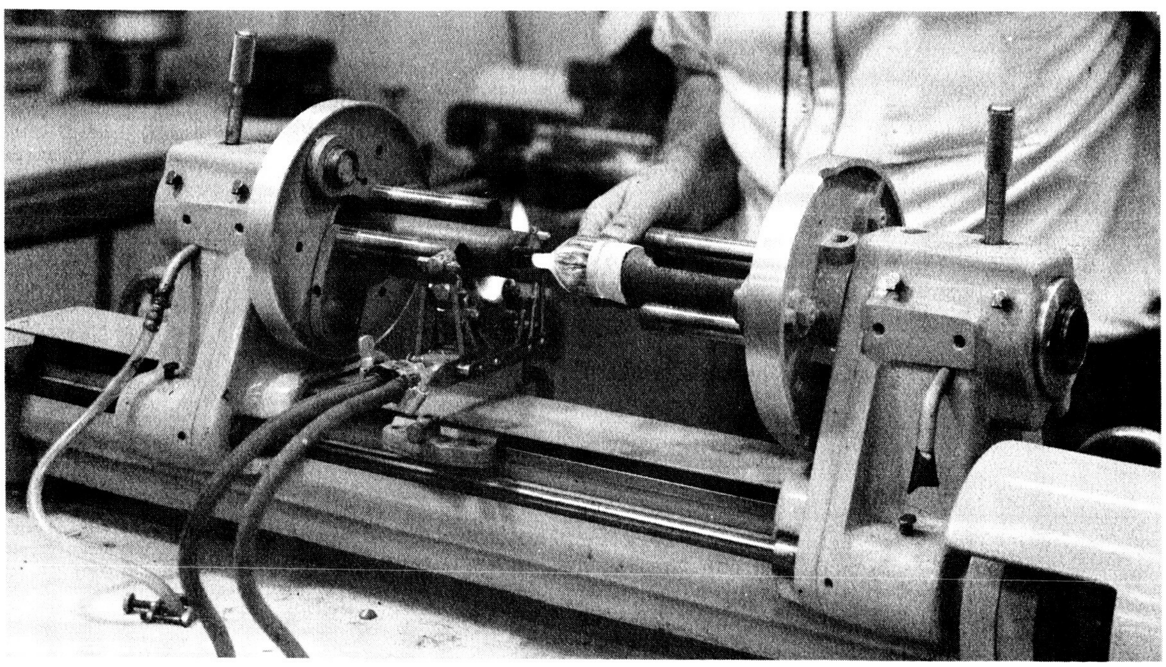


Figure 4. Uranium glass-tungsten electrode graded glass seals. The glass sheathed pins and the test chamber cap are heated in the glass blower's lathe, and then hot press formed into one piece.

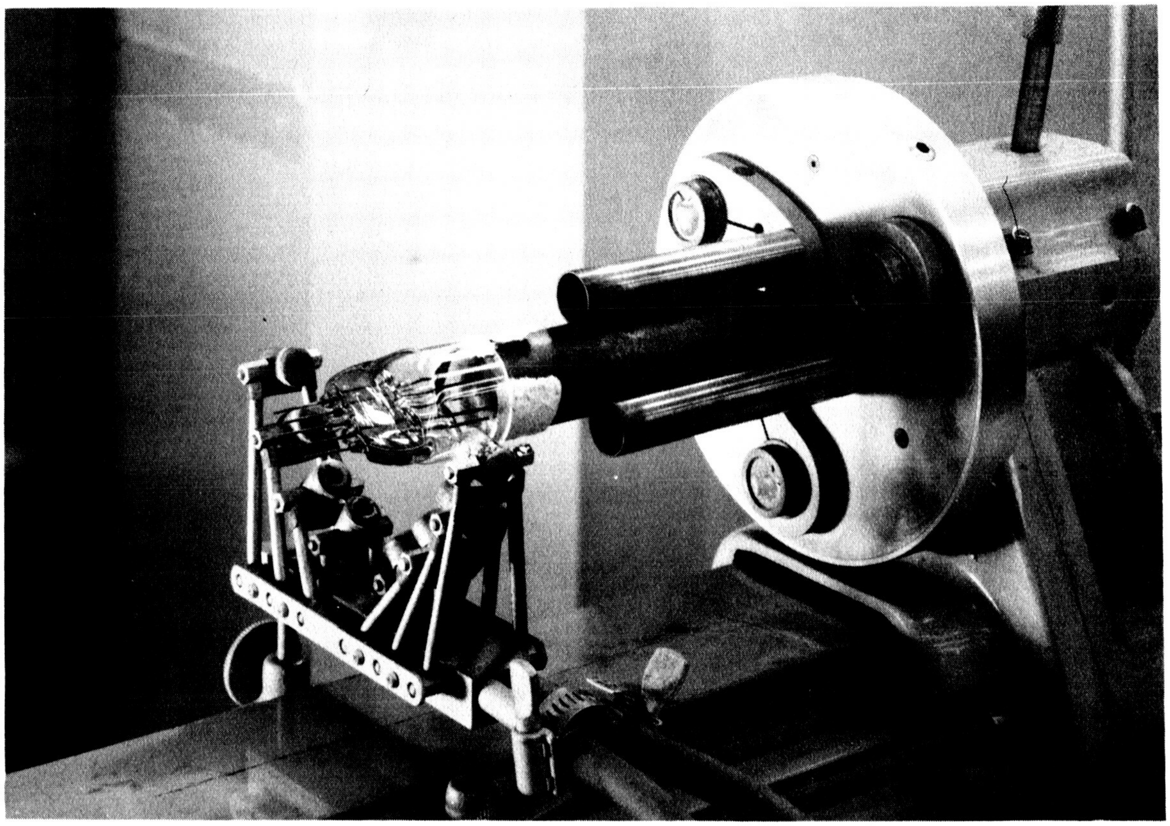


Figure 5. Completed uranium glass-tungsten electrode graded glass seal. Note how the pins are held in place before fusion by the graphite die.

As the illustrations cited show, provisions for spectroscopic analysis have been built into the test chamber. Infrared spectra of the resins and crosslinking agents have been taken for many of the materials which will be submitted to the test. Gases and condensible liquids from the cryotraps will be analyzed by infrared spectroscopy, and the spectrograms will be compared to those of the crosslinking agents and resins, taken earlier.

EXPERIMENTAL RESULTS OF PRELIMINARY SCREENING TESTS

The materials' names, types, manufacturers, hardener concentrations, cure schedules, and special considerations are shown in Table 1, as they were prepared for the preliminary screening tests. The numbers used are consistent with those used in earlier reports on this program.[7] The cures on the additional samples of Sylgard 182 were suggested by project management in Huntsville.

Table 2 summarizes the moisture resistance data for Shell Epon X24 and for the three alternate cures suggested for Sylgard 182. The numbering system for Table 1 is also used in Table 2.

The results of the Heat Aging Screening Test are summarized in Table 3 for the alternate cures of Sylgard 182, and for Shell Epon H25 and X24. The specimens are numbered according to the scheme used in Table 1.

The recovery of the materials from the visible effects of the moisture exposure is shown in Table 4. The data reported is for all materials not covered in previous reports, except LTV 615. Additional samples of this material were delayed in transit, and therefore the test could not be included in this report.

The cured Sylgard 182 produced according to the cures suggested by project management at Huntsville, and those materials reported in earlier reports on this project[7] are very close in their behavior. Figure 6 is a plot of all the data taken to date on Sylgard 182. The convergence of the data is very obvious.

Number	Material Name	Material Type	Material Manufacturer	Hardener Concentration	Cure Schedule	Special Considerations and Comments
9A	Sylgard 182	Unfilled silicone	Dow Corning	10 parts by weight Sylgard 182 cross-linking agent to 100 parts by weight resin.	8 hours at 100°C	Cures to a straw yellow trans-parent resin.
9B	Sylgard 182	Unfilled silicone	Dow Corning	10 parts by weight Sylgard 182 cross-linking agent to 100 parts by weight resin.	20 hours at 100°C	Cures to a straw yellow trans-parent resin.
9C	Sylgard 182	Unfilled silicone	Dow Corning	10 parts by weight Sylgard 182 cross-linking agent to 100 parts by weight resin.	4 hours at 150°C	Cures to a straw yellow trans-parent resin.
25	H-25	Unfilled epoxy resin	Shell Chemical	19 parts by weight Hughes Hardener "A" to 100 parts by weight of H-25.	4 hours at 20°C 16 hours at 105°C 24 hours at 125°C	Cures to yellow amber and trans-parent material.
26	X-24	Unfilled epoxy resin	Shell Chemical	16 parts by weight Hughes Hardener "A" to 100 parts by weight resin.	4 hours at 20°C 16 hours at 105°C 24 hours at 155°C	Cures to light yellow amber transparent material. Base resin is pure diglycidyl ether of bisphenol A.

Table 1. Material summary.

Sample Number	Sample	Cure Schedule	Curing Agent	Average Weight Change, percent	Durometer Before	Durometer After	Passes Test	Comments on Appearance
9A	Dow Corning Sylgard 182	8 hours at 100°C	Sylgard 182 Curing Agent	+0. 21	$42A_2$	$52A_2$	Yes	During exposure material turns to a translucent light yellow material
9B	Dow Corning Sylgard 182	20 hours at 100°C	Sylgard 182 Curing Agent	+0. 27	$41A_2$	$52A_2$	Yes	During exposure material turns to a translucent light yellow material
9C	Dow Corning Sylgard 182	4 hours at 150°C	Sylgard 182 Curing Agent	+0. 21	$44A_2$	$52A_2$	Yes	During exposure material turns to a translucent light yellow material
26	Shell X-24	4 hours, 20°C 16 hours, 105°C 24 hours, 155°C	Hughes Curing Agent A	+0. 98	89D	89D	Yes	No visible change

Table 2. Summary of moisture resistance tests.

Material Number	Material and Manufacturer	Curing Agent	Cure Schedule	Percent Weight Loss, hours	Shore Hardness Before	Shore Hardness After	Passes Test	Conditional Pass	Comments
9A	Sylgard 182 Dow Corning	Sylgard 182 Curing Agent	8 hours at 100°C	-1.1 545 hours	$40A_2$	$54A_2$	Yes		Yellow color darkened in test
9B	Sylgard 182 Dow Corning	Sylgard 182 Curing Agent	20 hours at 100°C	-1.1 545 hours	$41A_2$	$54A_2$	Yes		Yellow color darkened in test
9C	Sylgard 182 Dow Corning	Sylgard 182 Curing Agent	4 hours at 150°C	-0.97 545 hours	$43A_2$	$54A_2$	Yes		Yellow color darkened in test
25	H-25 Shell Chemical	Hughes Hardener "A"	4 hours, 20°C 16 hours, 105°C 24 hours, 125°C	-0.29 504 hours	86D	90D	Yes		Material turns opaque and dark
26	X-24 Shell Chemical	Hughes Hardener "A"	4 hours, 20°C 16 hours, 105°C 24 hours, 155°C	-0.31 545 hours	89D	89D	Yes		Material turns opaque and dark

Table 3. Summary of heat aging.

Material Number	Material	Original Appearance	Post Moisture Test	Post Recovery Bake	Thermal Resistance Weight Change, percent	Moisture Resistance Weight Change, percent	Recovery Bake Weight Change, percent
8	Sylgard-182	Water white, clear	White, translucent	Yellow, transparent	-0.26	+0.70	-0.92
9	Sylgard-182	Water white, clear	White, translucent	Yellow, transparent	-0.45	+0.89	-0.90
9A	Sylgard-182	Yellow, clear	Yellow, translucent	Clear, slightly more yellow	-1.10	+0.21	-0.80
9B	Sylgard-182	Yellow, clear	Yellow, translucent	Clear, slightly more yellow	-1.10	+0.27	-0.84
9C	Sylgard-182	Yellow, clear	Yellow, translucent	Clear, slightly more yellow	-0.97	+0.21	-0.75
10	XR-6-3477	Water white, clear	Whitish, translucent	Returned to original appearance	-0.74	+0.03	-0.50
13	LTV-615	Water white, clear	White, translucent	No data available yet	-1.05	–	–
14	LTV-615	Water white, clear	White, translucent	No data available yet	-1.05	–	–

Table 4. Recovery from visible effects of moisture resistance tests.

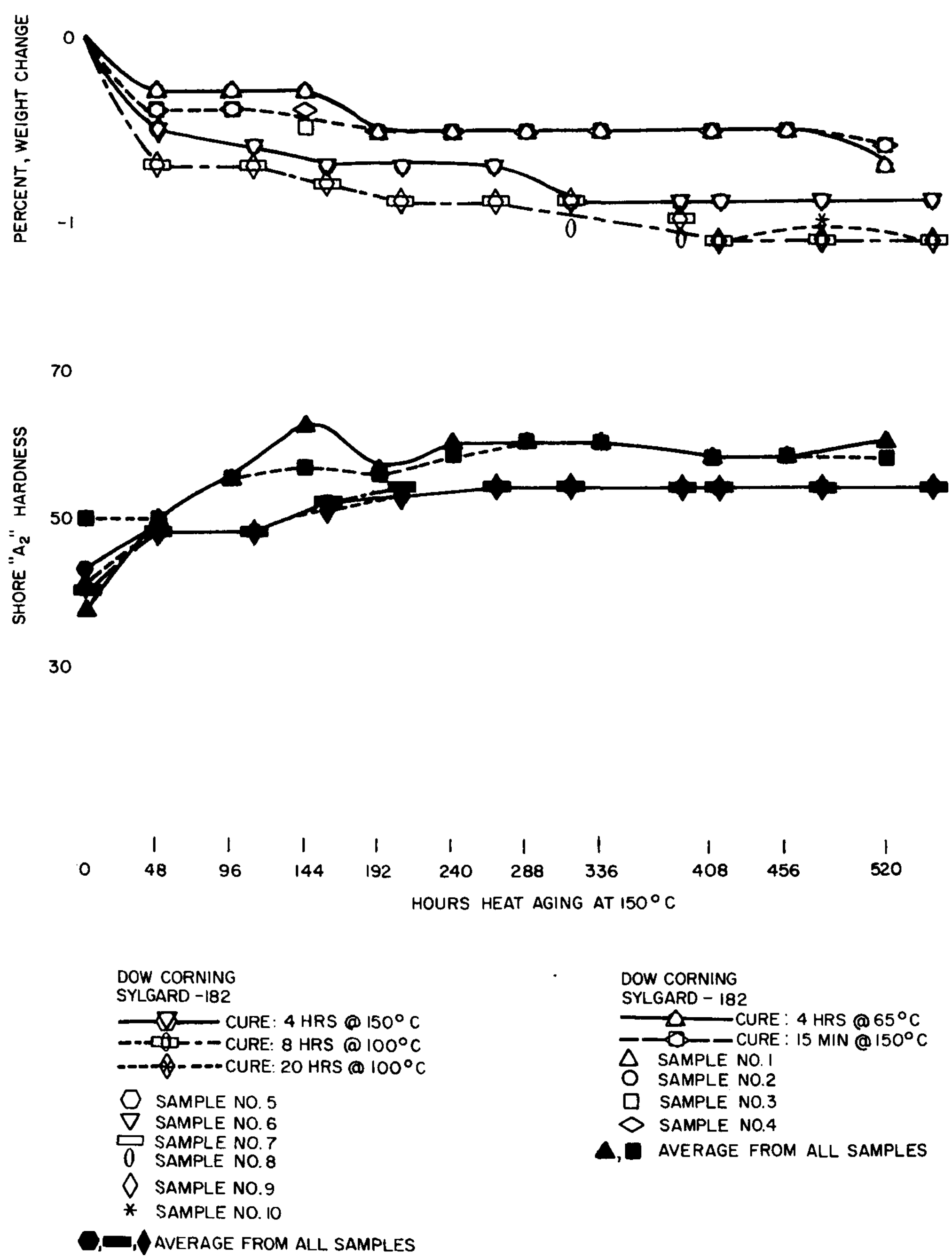


Figure 6. Effect of thermal stress on Sylgard-182 as a function of cure, hardness and weight loss. The curves are almost identical, the major difference being in the hardness. The cure schedules apparently cure the resin to the same degree.

Figure 7 shows the final results for Epon H25 behavior under thermal stress. The color of the material becomes dark enough to render it opaque after the 500-hour exposure to heat. This is due to oxidation of the aromatic amine crosslinked epoxy and the generation of dark colored dye bodies from the amine.

The behavior of the Epon X24 is shown in Figure 8. The curing ratio of 16 parts of hardener to 100 parts of resin was suggested by the resin supplier. Work on this material in other programs indicates that 20 parts of hardener to 100 parts of resin seems to give a more satisfactory cured resin. As in the case of Epon H25, the X24 material darkened to the point of opacity during the 545 hours of thermal stress.

PRELIMINARY EVALUATION TESTS

As the materials which passed the Preliminary Screening Test were passed along to the Preliminary Evaluation phase of the program, infrared spectra of the prepolymers and the crosslinking agents were taken. These spectra were taken to elucidate the general structure of the polymers, provide clues to possible synthesis solutions to the requirements of the programs, and to determine which constituents of the cured polymer contribute the most materials to any volatile constituents which might be cryotrapped during the Preliminary Evaluation phase of the program.

An analysis of the infrared findings on the sample materials, along with representations of the spectral traces, follows.

Dow Corning XR-6-3477, Sylgard 182, General Electric 155-45-3026, LTV 615, S-5364, and Minnesota Mining and Manufacturing SK-495 produced very similar spectra (see Figures 9 through 14). Their spectral characteristics indicate that they are principally methyl silicones with trimethyl end groups. All contain a small amount of Si-OH groups. General Electric LTV 615 appears to have the longest chain length as indicated by the lowest Si $(CH_3)_3$:Si (CH_3) absorbance ratio of the group.

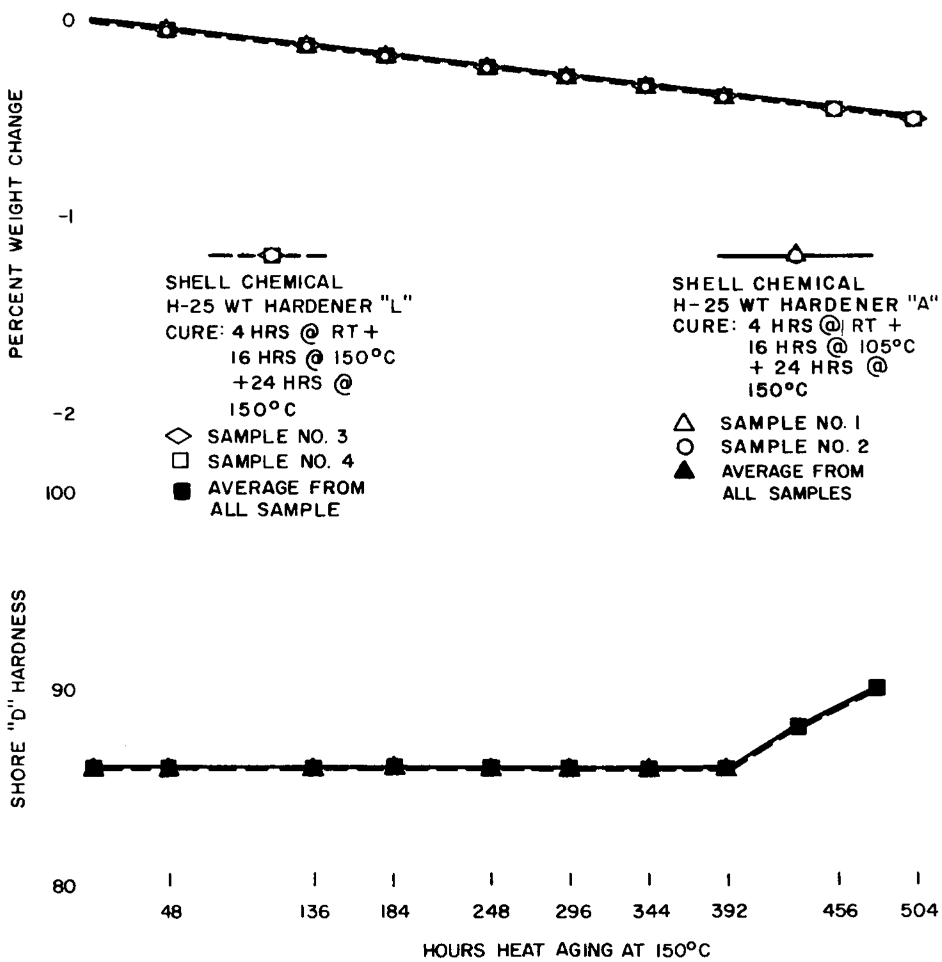


Figure 7. Shell Chemical Corporation H-25 hardness and weight change under thermal stress. The material shows very small changes in hardness and weight out to 504 hours.

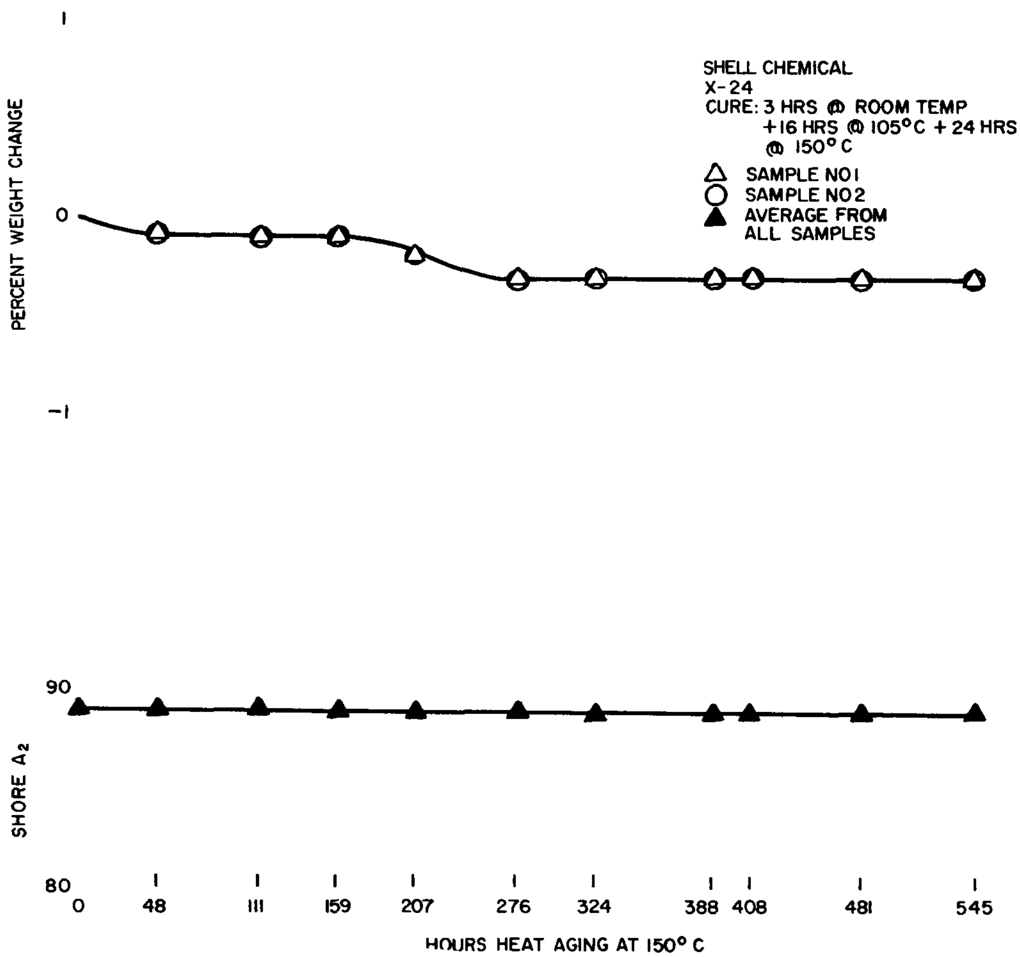


Figure 8. Shell Chemical Corporation X-24 resin hardness and weight change under thermal stress. The material shows no changes in hardness and weight out to 545 hours.

Figure 9. Infrared spectrum of Dow Corning XR-6-3477 resin.

Figure 10. Infrared spectrum of Dow Corning Sylgard 182 resin.

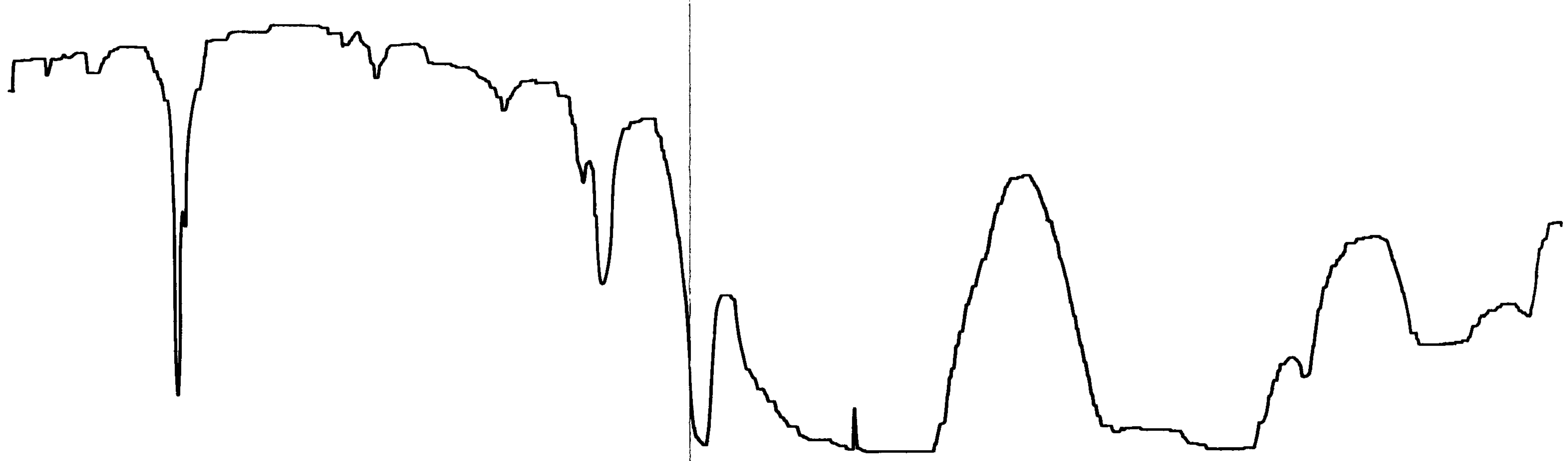


Figure 11. Infrared spectrum of General Electric 155-3026 resin.

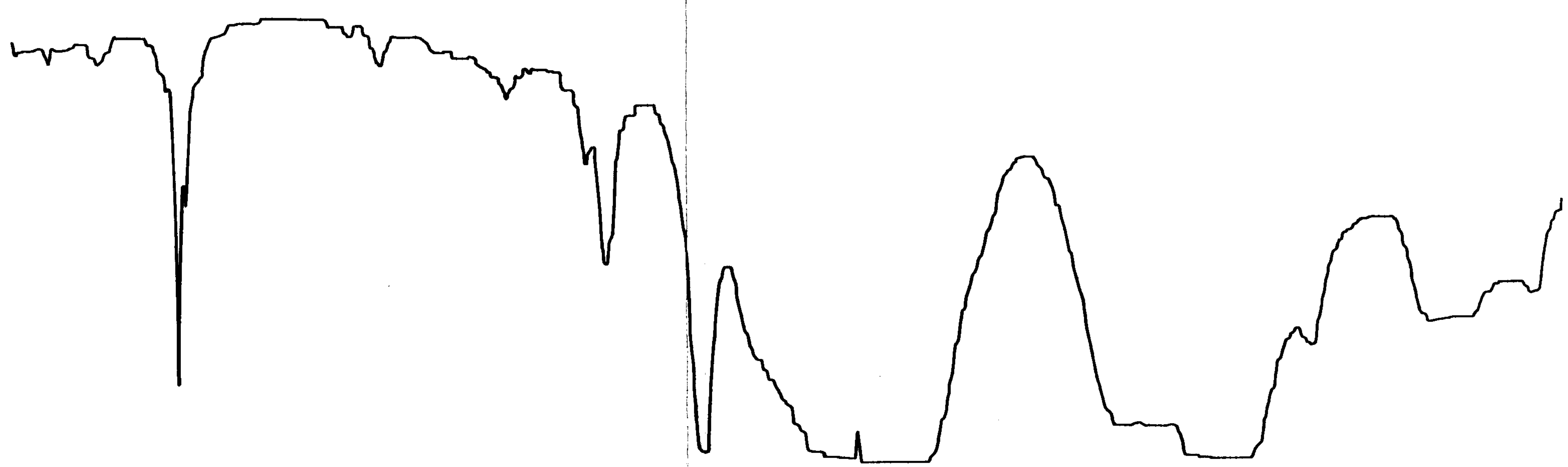


Figure 12. Infrared spectrum of General Electric LTV615 resin.

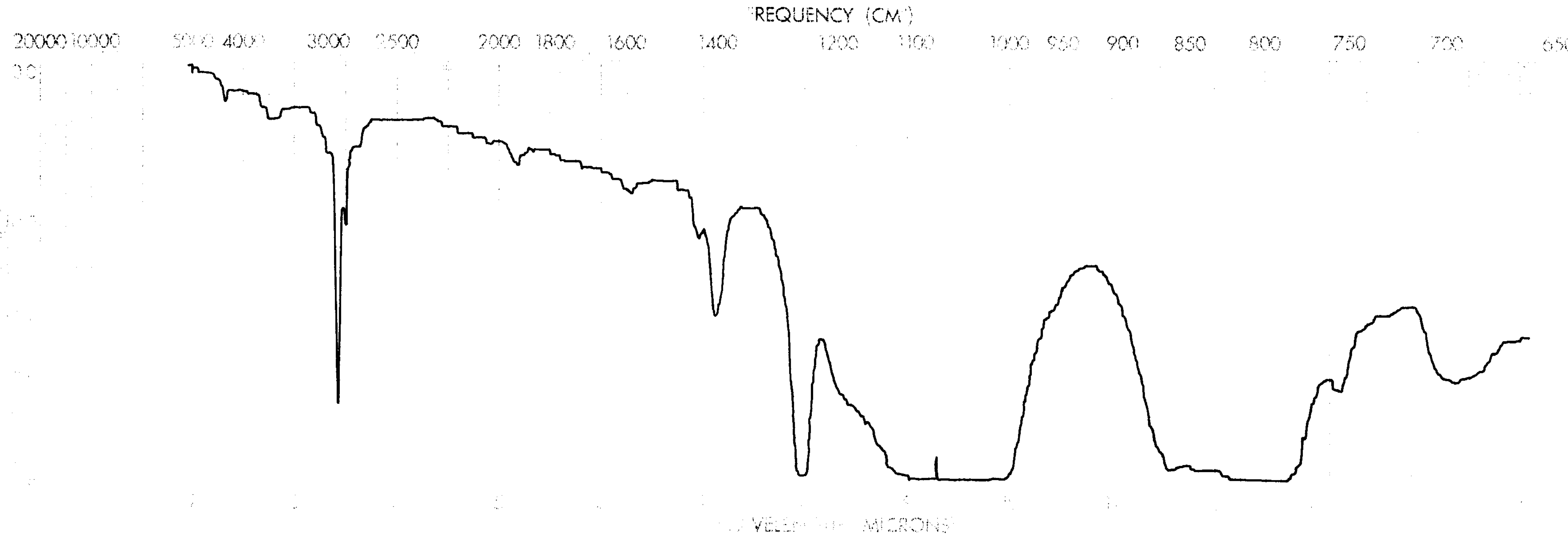


Figure 13. Infrared spectrum of General Electric S-5364 resin.

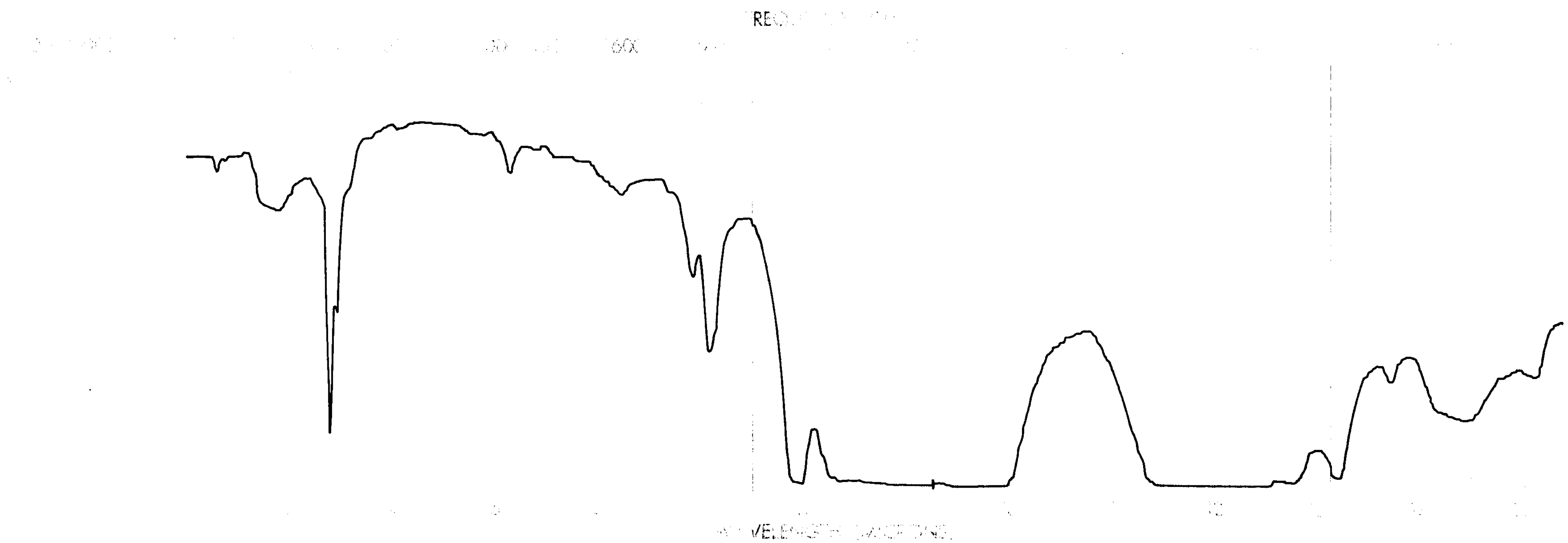


Figure 14. Infrared spectrum of Minnesota Mining and Manufacturing SK 495 resin.

Minnesota Mining and Manufacturing SK 493 and SK 496 (see Figures 15 and 16) contain phenyl as well as methyl groups on the siloxane chain. They also possess trimethyl terminal groups.

The spectra of the curing agents for Dow Corning Sylgard 182, XR-6-3477, General Electric 155-45-3026, LTV 615, and S-5364, are characterized by the presence of the Si-H band at 2230 cm^{-1} (see Figures 17 through 21). S-5364 crosslinking agent appears to have the largest percentage of Si-H groups per molecule. All show Si-O-Si, and $Si\text{-}CH_3$ vibrations. Sylgard 182 curing agent also contains Si  groups.

The spectra of the crosslinking agents for Minnesota Mining and Manufacturing SK 493 and SK 495 indicate short chain silanols possessing ethylenic groups. That of SK 496 also indicates an ethylene type siloxane but silanol groups are practically absent. The spectra fail to show a Si-C band at 1250-60 cm^{-1} or $Si\text{-}(CH_3)_3$ bands at 841 cm^{-1} and 754-6 cm^{-1}. This indicates that the C groups are not attached directly to Si but probably through O linkages (see Figures 22, 23 and 24).

R-7521 and R-7501 are both phenyl methyl silicones with very similar structures. The amount of phenyl groups in these structures is quite high. Apparently no trimethyl terminal groups are present as indicated by the absence of $Si(CH_3)_3$ bands at 855 cm^{-1} and 755 cm^{-1}. The infrared spectral pattern between 2000 cm^{-1} and 1600 cm^{-1} indicate monosubstituted phenyl groups in the compounds. Molecular still treatment of R-7501 to produce R-7501 H apparently resulted in removal of low molecular weight fractions (see Figures 25 and 26). The spectra of the curing agents for these resins are straightforward. Disilylbenzene shows the characteristic Si-H band at 2230 cm^{-1}. An infrared pattern indicative of parasubstituted phenyl groups is present between 2000 cm^{-1} and 1600 cm^{-1}. The spectrum of dicumyl peroxide shows the monosubstitution pattern for phenyl groups between 2000 cm^{-1} and 1600 cm^{-1}, monosubstitution bands at 1490 cm^{-1}, 1175 cm^{-1}, 1160 cm^{-1}, 1030 cm^{-1}, 905 cm^{-1}, 760 cm^{-1}, 690 cm^{-1}, and bands due to the isopropyl groups at 1370 cm^{-1}, 1360 cm^{-1}, and 1150 cm^{-1}.

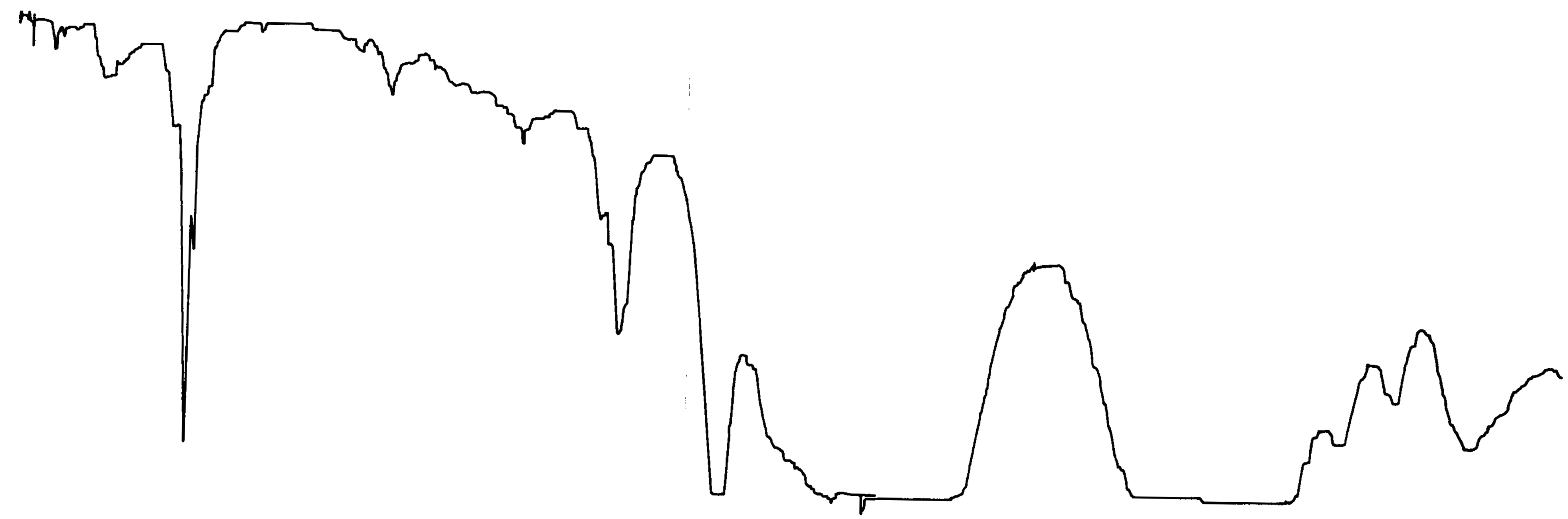


Figure 15. Infrared spectrum of Minnesota Mining and Manufacturing SK 493 resin.

Figure 16. Infrared spectrum of Minnesota Mining and Manufacturing SK 496 resin.

Figure 17. Infrared spectra of Dow Corning Sylgard 182 crosslinking agent.

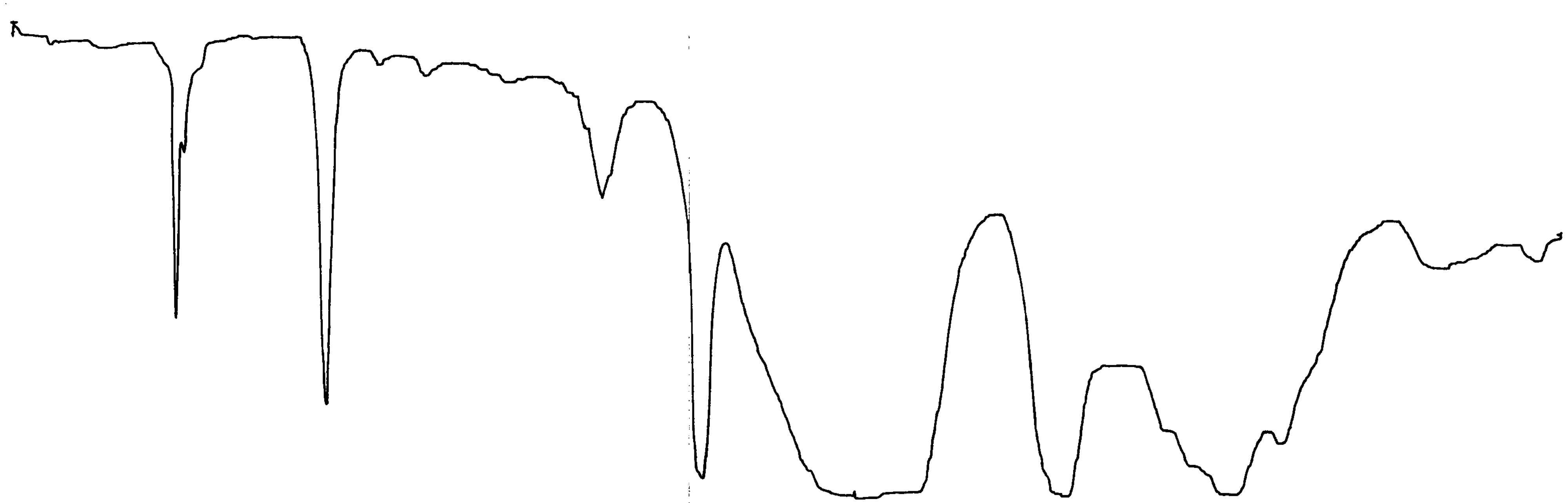


Figure 18. Infrared spectrum of General Electric LTV 615 crosslinking agent.

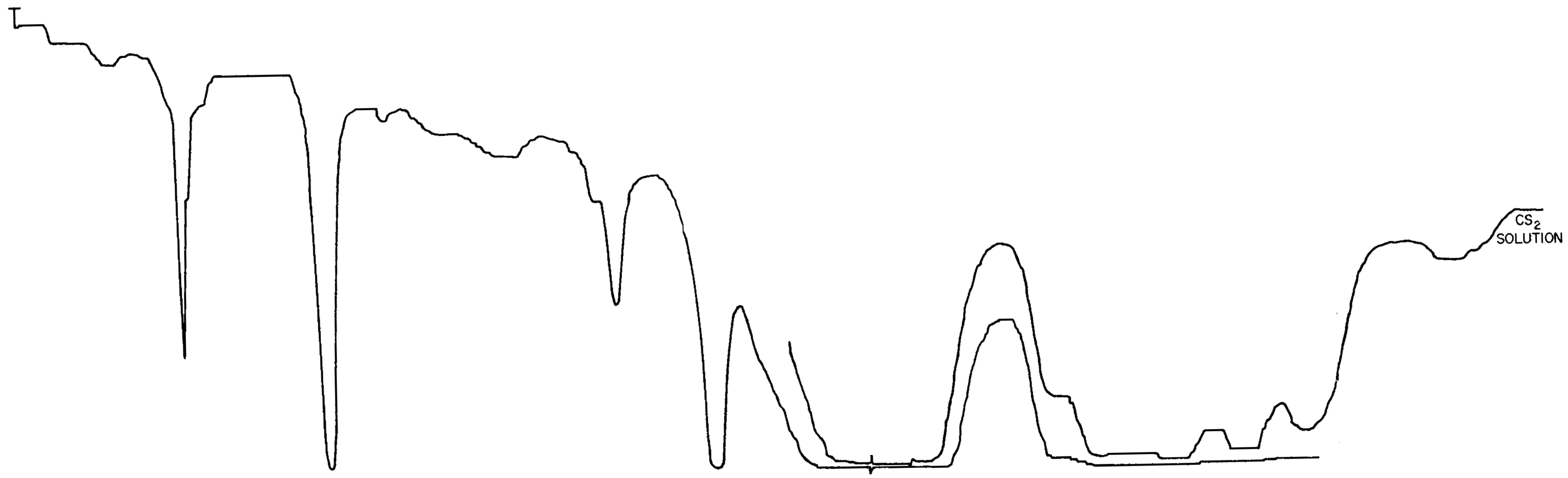


Figure 19. Infrared spectrum of Dow Corning S-5364 crosslinking agent.

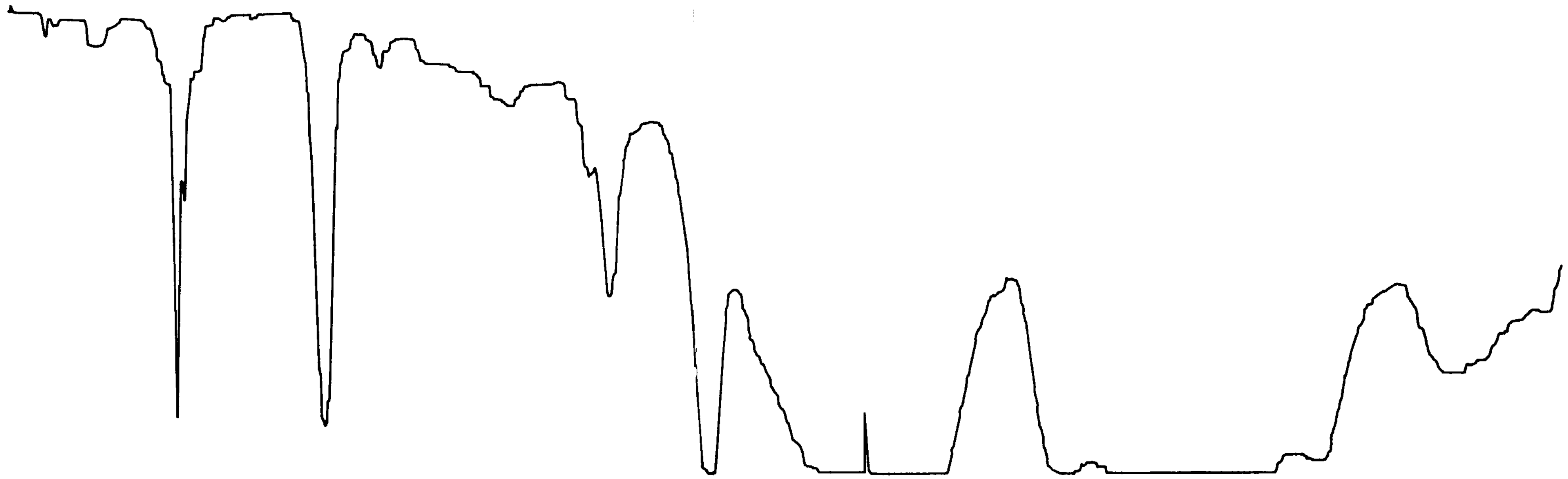
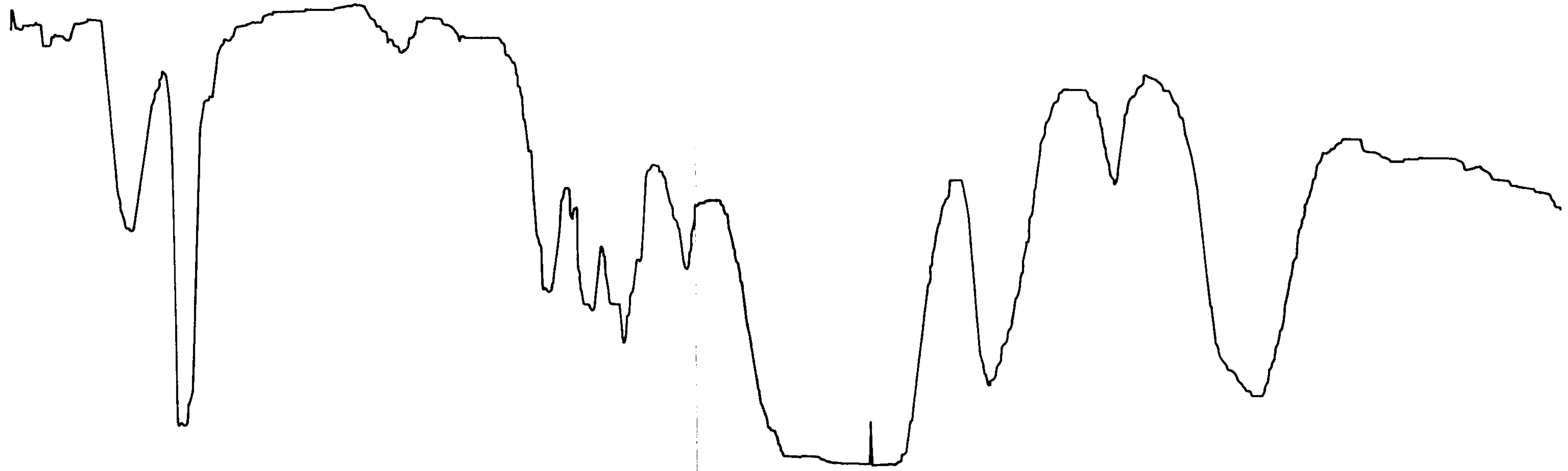


Figure 21. Infrared spectrum of the crosslinking agent for Dow Corning XR-6-3477.



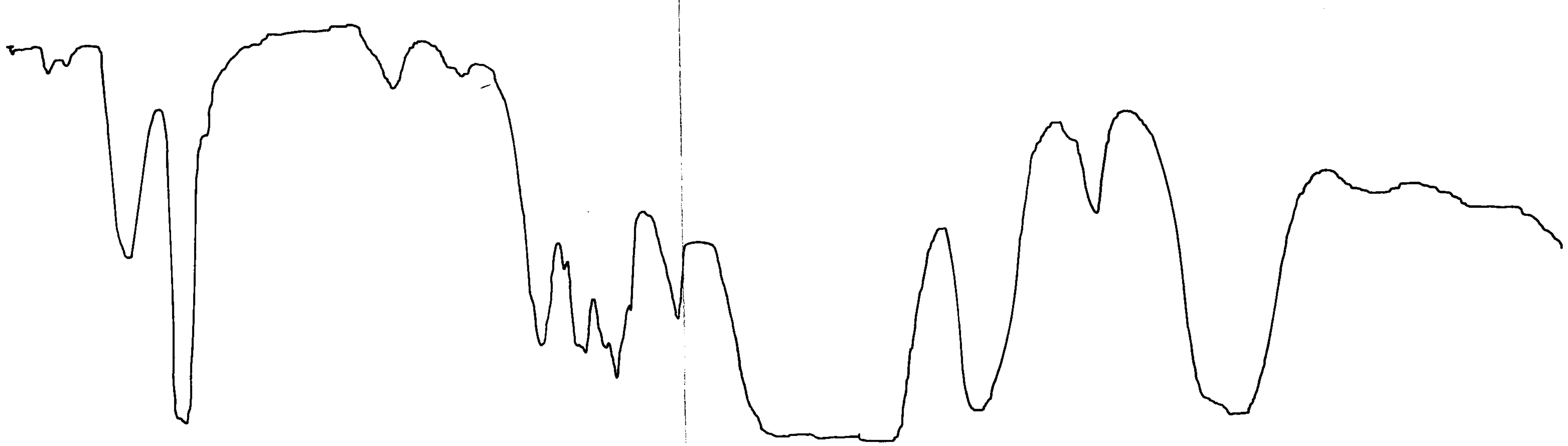


Figure 23. Infrared spectrum of crosslinking agent for Minnesota Mining and Manufacturing SK 495 resin.

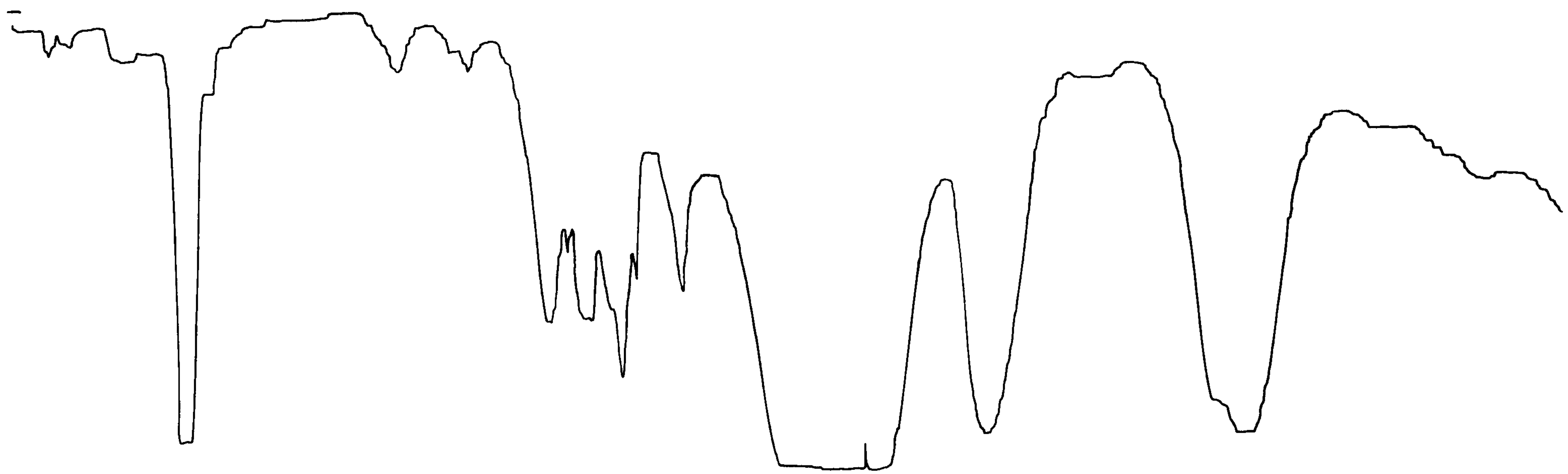


Figure 24. Infrared spectrum of crosslinking agent for Minnesota Mining and Manufacturing SK 496 resin.

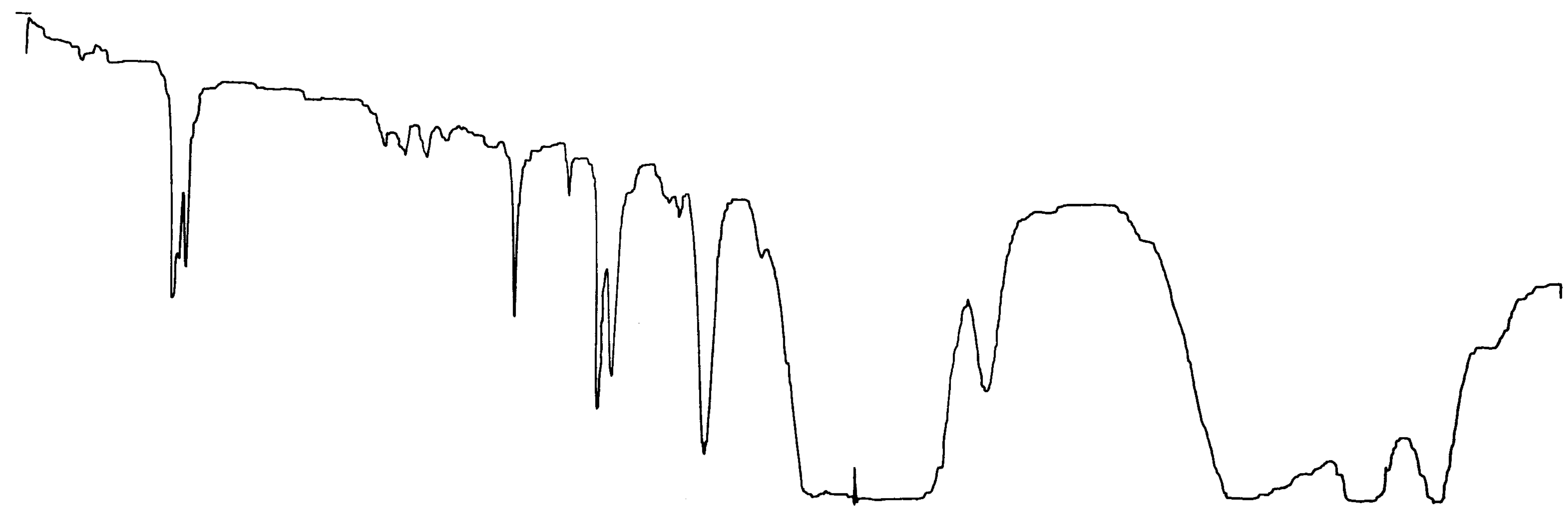


Figure 25. Infrared spectrum of Dow Corning R-7501 resin.

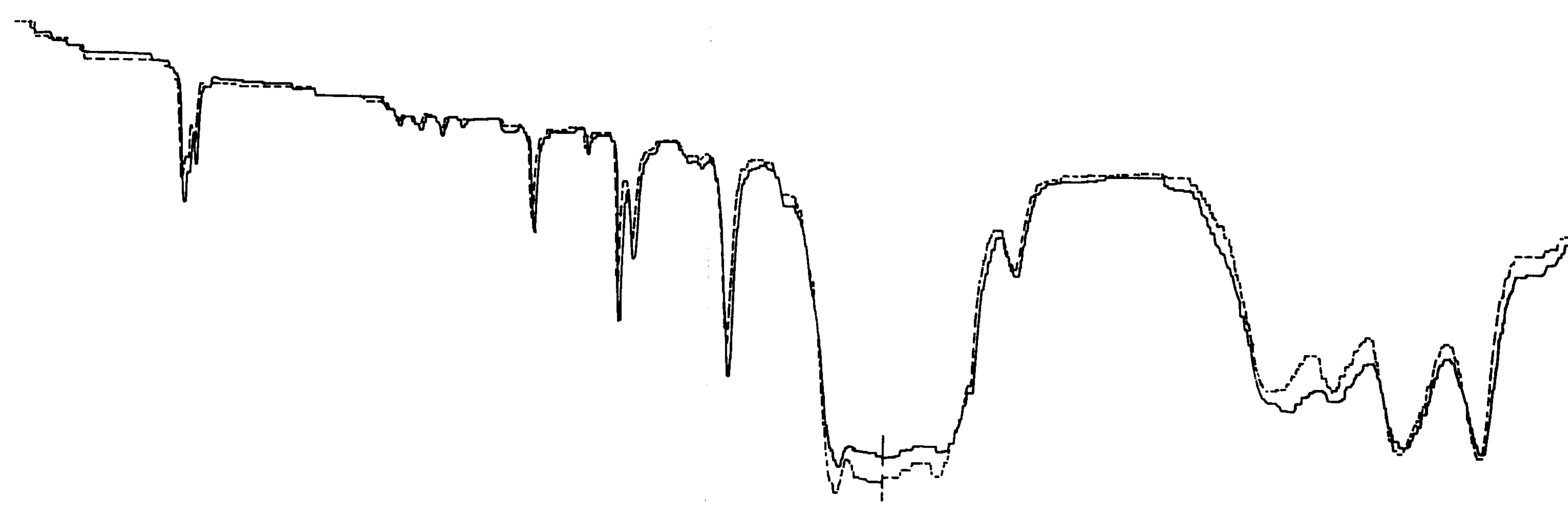


Figure 26. Infrared spectra of Dow Corning R-7501 resin and molecularly distilled R-7501 coded as R-7501-H.

Probable assignment for the peroxide structure is given to the band at 849 cm^{-1} (see Figures 27 and 28).

H25 and X24 resins are epoxy compounds. Epoxy groups are shown by the bands at 913 cm^{-1} and 1240 cm^{-1} in the infrared spectrum of these materials. A small amount of -OH groups is present in H25, as indicated by the band at 3450 cm^{-1}. Both resins have quite low epoxide equivalents, below 200, as determined by the relative intensities of the epoxide bands at 913 cm^{-1}.[8] The epoxide equivalent of H25 is slightly higher than that of X24.

The spectrum of the curing agent for the epoxy resins, Hardener A, shows bands for -SH group at 2480 cm^{-1} and for $-NH_2$ group at 3400 cm^{-1}, 3300 cm^{-1} and 1600 cm^{-1} (see Figures 29, 30 and 31).

POLYMER MODIFICATION

The disparity between the coefficients of linear thermal expansion specified by project management in Huntsville, and those that are obtainable from the unmodified materials being evaluated under the present program, makes it necessary to investigate the effect of fillers on the resins currently being evaluated.

Quartz, because of its low dielectric constant, high degree of transparency, and low coefficient of expansion, would seem most likely to fulfill the requirements of this program.

Some of the Hughes personnel whose time is partially committed to this program are engaged in improving the properties of resins in support of the Phoenix Fire Control System for the F-111A, and have discovered a mathematical relationship between the amount of resin, the amount of filler, and the magnitude of the resin-filler composite's coefficient of linear thermal expansion. This discovery will be used to help meet the goals of this program.

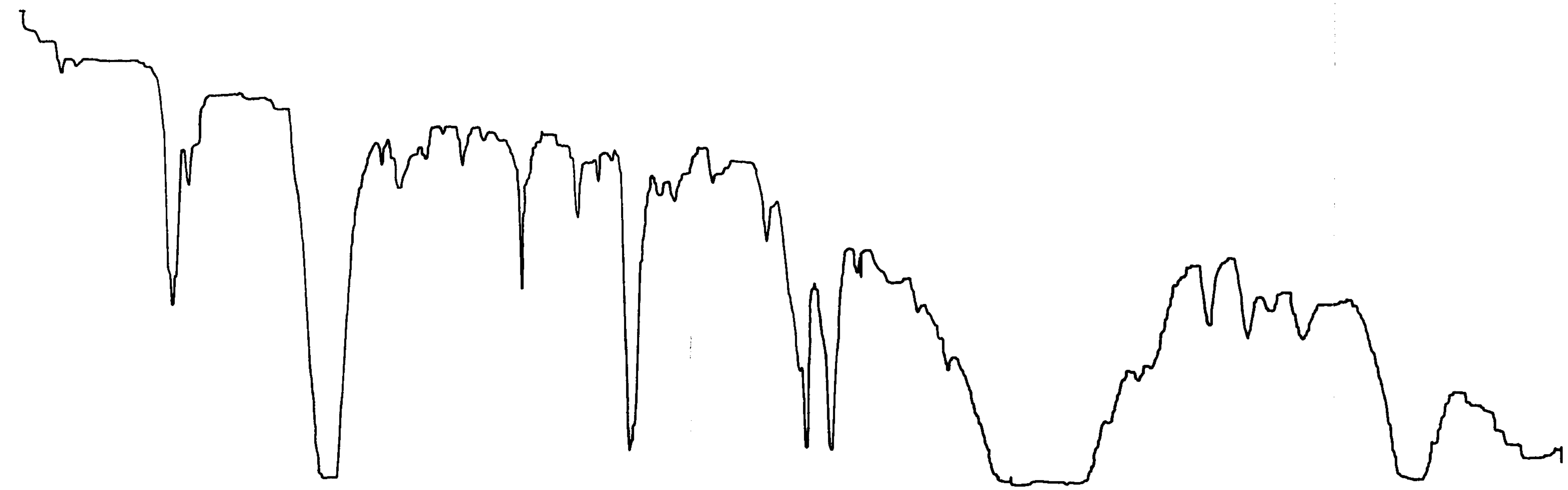


Figure 27. Infrared spectrum of disibyl benzene.

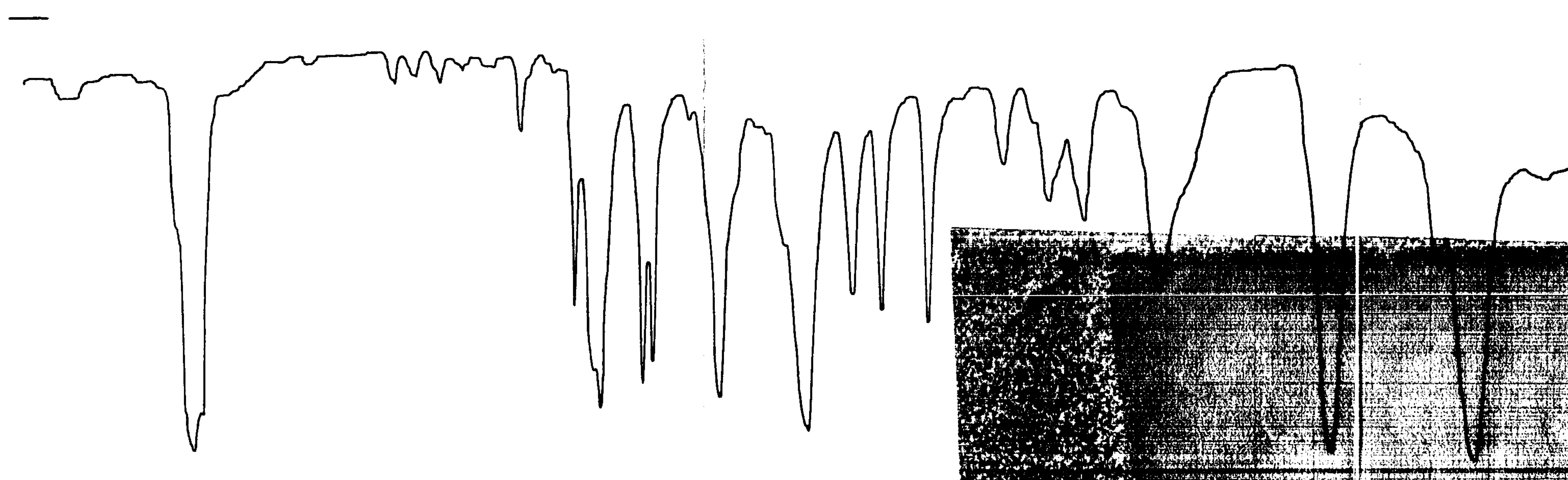


Figure 28. Infrared spectrum of dicumyl per

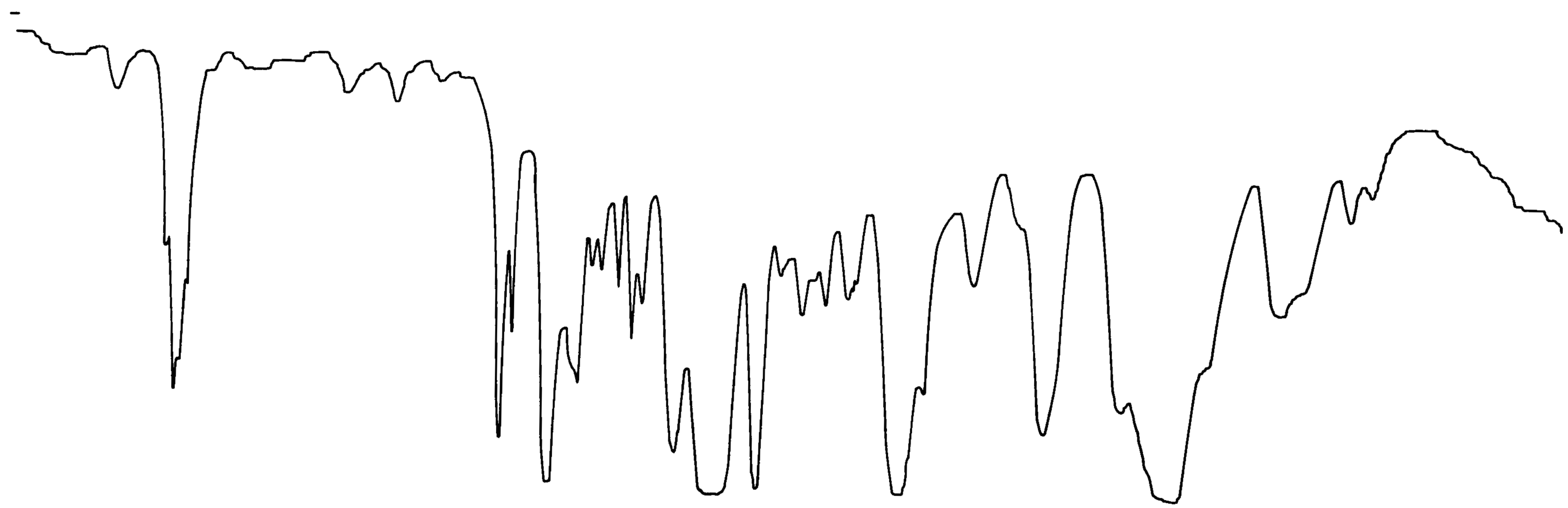


Figure 29. Infrared spectrum of Shell Epon H25 resin.

Figure 30. Infrared spectrum of Shell Epon X-24 resin.

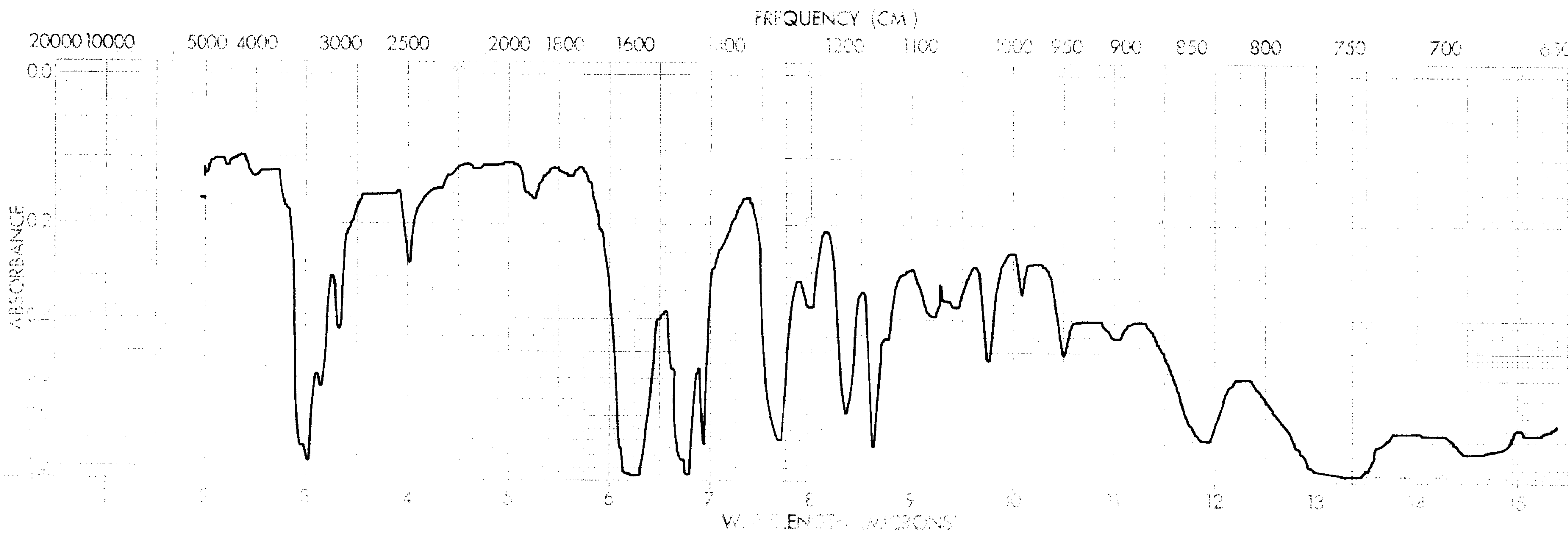


Figure 31. Infrared spectrum of Hughes Aircraft Company Hardener System A.

FILLER

Because of its excellent electrical properties, quartz has been found to be useful as a filler for such polymer systems as epoxy resins and silicone rubbers. Although chemically quartz is a relatively simple compound, SiO_2, it can have a number of physical crystalline forms. Some of these as well as their transition temperature and specific gravities are shown below.

$$\underset{2.65}{\text{Quartz}} \;\longleftarrow 870°\text{C} \longrightarrow\; \underset{2.26}{\text{Tridymite}} \;\longleftarrow 1470°\text{C} \longrightarrow\; \underset{2.32}{\text{Cristobalite}},\ \underset{1710°\text{C}}{\text{m. p.}}$$

The last two forms can exist indefinitely at room temperature, although at such a temperature they are metastable. Each of the above polymorphic forms of silica exists in two subsidiary forms with the following transition points:

α - β quartz	α - β tridymite	α - β cristobalite
573°C	120-160°C	200-275°C

Crystals of quartz, tridymite, and cristobalite consist of three-dimensional networks of tetrahedra of SiO_4 joined so that each oxygen atom is common to two tetrahedra. In quartz the tetrahedra are so linked that they acquire a spiral formation, and the crystal is optically active. The interior of quartz is in electrical equilibrium. However, the surface of the crystals is composed of ions whose valences are not completely satisfied. This allows volatile contaminants such as water to be absorbed on the surface and held by hydrogen bonding. A schematic representation is shown in the diagram below.

$$\underbrace{-\,O\,-\,\overset{\overset{H_2O}{\downarrow}}{Si}\,-\,O\,-\,\overset{\overset{H_2O}{\downarrow}}{Si}\,-}_{\text{crystal surface}}$$

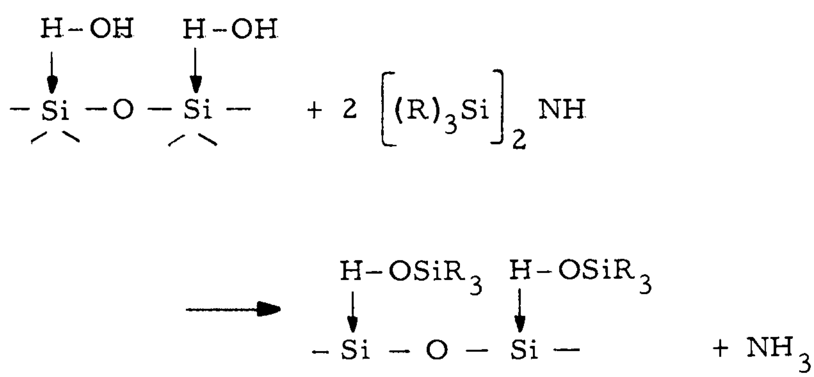
The energy of bonding is roughly 2 to 5 Kcal/mol. When such quartz is used as a filler, outgassing of the water may occur under certain conditions which may adversely affect the properties of the composite. This water can be removed by calcination. Such a calcined quartz flour has been used as fillers for epoxy casting resins for electrical applications.[9]

Fissures, cracks and irregularities in the quartz particles may also represent some serious problems in that it greatly increases the surface area where absorption can take place and also makes it difficult to outgas the quartz before being used as a filler. Improperly outgassed quartz could lead to the evolution of undesirable volatiles during operating conditions to which the composite system may be subjected.

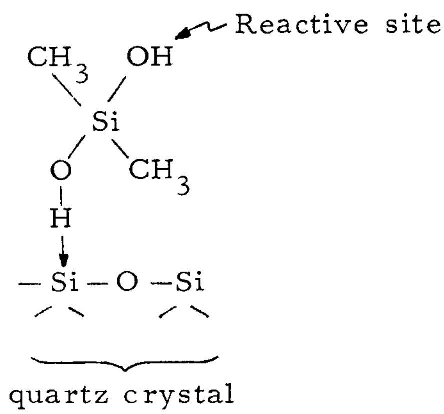
Thus, the chief problems which may be present in the use of quartz as a filler may be summarized as (1) the highly polar nature of the surface of the crystal which allows the attraction of active volatile species and (2) the physical characteristics of the surface which may lead to inability to outgas the quartz properly. Below are listed a number of procedures which improve the compatibility of quartz with epoxy and silicone resin systems.

1. <u>Replacement of Adsorbed Water with Compounds that are Compatible with the Resin Systems</u>

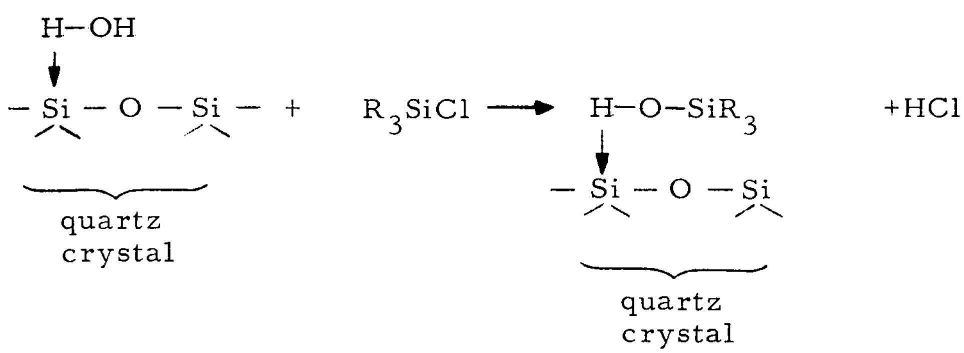
a. <u>Reaction with Organosilazanes</u>



Silizanes such as hexamethylcyclotrisilazane may be advantageously employed since they offer greater possibilities of additional bonding between the polymer and the quartz crystal. For example, one can envision the following layer on the quartz.



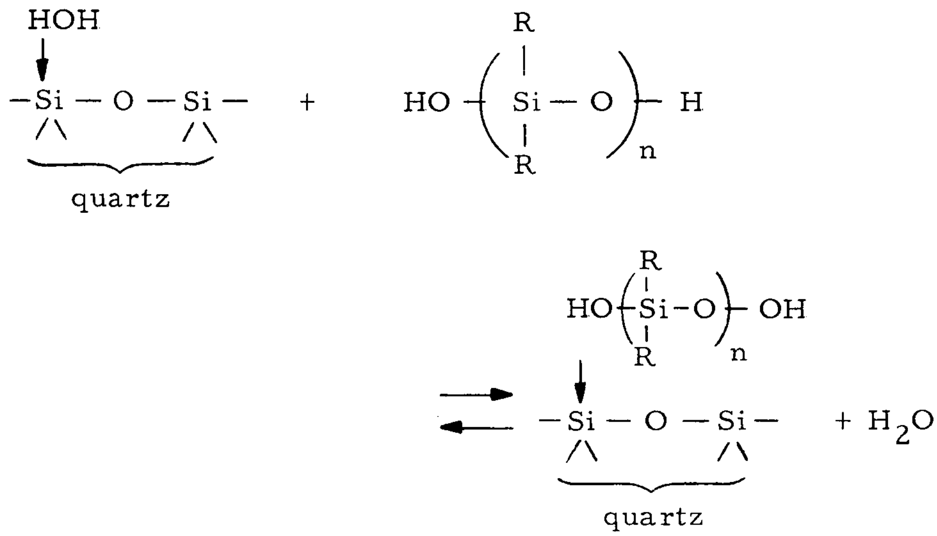
b. <u>Reaction with Organohalosilanes</u>



Di and trifunctional silanes can also be employed to give additional bonding between the quartz substrate and the polymer system.

c. <u>Replacement of the Water with Silandiols</u>

By equilibrating quartz with a silandiol it is possible to replace some of the adsorbed water. If the molecular weight of the silandiol is large enough, outgassing would be negligible. For example, the following equilibrium is possible:



The residual hydroxy groups can then enter into combination with the polymeric system.

2. Removal of Water by Physical Means

It is possible to remove the water by heating and the simultaneous application of a vacuum so that the outgassed material is removed from the system. The resulting clean surface is then exposed to a silicone monomer or epoxy monomer which can then act as an effective substrate to bond the polymer to the quartz.

3. Modification of the Surface of the Quartz to Eliminate Cracks and Ridges

The purpose of this procedure is to minimize the surface area with respect to volume. One method by which this can be accomplished is through the use of $HF-N_2$ mixture. The sharp ridges would be more reactive and this action would result in a smoothing action. The HF would react with the SiO_2 according to the following equation.

$$SiO_2 + 4HF \longrightarrow SiF_4(gas) + H_2O .$$

The resulting treated silica would then be heated and outgassed.

PREDICTING COEFFICIENT OF LINEAR THERMAL EXPANSION

Several times in the past attempts have been made to predict the coefficient of thermal expansion of a resin-filler mixture from a knowledge of its composition and of the coefficients of its ingredients. The value of such predictions for screening ingredients and for formulating materials to match given coefficients is quite obvious.

If the filler is inert with respect to the resin one would expect the coefficient of the mixture to have some value intermediate between those of the ingredients, and such is the case. Thus, the problem is that of properly weighting the coefficients of the ingredients. Obviously the weighting factors should be such as to allow for both the volumes of the ingredients and the shapes of the filler particles.

In the past the difficulties involved in allowing for the shape factor led to the development of two formulae, one for finely divided fillers and one for fibrous fillers. A relatively large amount of data on elastomers loaded with different finely divided fillers was available for checking one of the formulae. A statistical analysis comparing the observed values with those calculated from the formula indicated that the discrepancies were not due to random error but rather that there was a definite positive bias to the calculated values.

Recently in the course of some work on a loaded epoxy resin, the same problem was encountered. This time attempts made to remove the bias were successful. It was found that two modifications of the original model were necessary to accomplish this. There were (1) use of geometric averaging rather than arithmetic, (2) both the volume weighting factor and the coefficient of the filler should be those of the zero porosity filler.

Since neither of the fillers used in this work had zero porosity, the latter condition might appear to violate physical principles. However, when one considers that the thermal expansion of a hollow sphere is the same as that of a solid sphere of the same material and the same radius, then the logic of the condition is plain. A porous mixture should have the same coefficient of thermal expansion as a non-porous mass of the same composition.

The relationship would make screening of resin-filler easy. The upper limit of the coefficient of a given system will be the coefficient of the continuous phase (resin). The lower limit will be dependent upon the coefficients of both the continuous and the disperse phase and also upon the closest packing ratio. By assuming that the filler particles consist of small uniform spheres, a reasonable approximation to the ratio can be obtained. Thus, the condition for obtaining a specified coefficient from a given system is:

$$\alpha_r^{0.26} \alpha_f^{0.74} < \alpha < \alpha_r$$

where

α = the specified coefficient
α_r = resin coefficient
α_f = filler coefficient

If this condition is met, then the material having the desired coefficient of thermal expansion can be formulated by using a resin-volume fraction of:

$$v_r = \log(\alpha/\alpha_f)/\log(\alpha_r/\alpha_f)$$

The filler volume fraction is:

$$v_f = 1 - v_r$$

CONCLUSIONS

At the present time work has been completed or is going forward in five of the six phases of the program. Present indications are that the program will produce a formulation or formulations based on commercially available materials that will have a dielectric constant between 4 and 5 when measured at 1000 cycles, and a coefficient of linear thermal expansion that will approach steel, and will stand prolonged service at 150° C with 24 hour exposure to 200° C. At the present time the hardness, adhesion, transparency, elongation, and cure schedules are subject to doubt.

FUTURE WORK

The preliminary evaluation tests will be completed during the month of May 1964. The tests are scheduled to start sometime before 1 May 1964.

The coefficient of linear thermal expansion will be investigated, and all efforts will be made to improve this property significantly in the time remaining to the program.

The vendors who supplied materials for evaluation in this program will be contacted if their materials reached the preliminary evaluation phase of the program. The results of this vendor liaison will be used to guide any future work with commercial and semi-commercial materials, and to give some direction to any future synthesis program related to this project or its extensions.

APPENDIX A

REFERENCES

1. Development of Improved Heat Sterilizable Potting Compounds, R. B. Feuchtbaum, NAS 8-5499, Hughes Aircraft Company, Report P63-61, Culver City, California, 15 October 1963.

2. Request for Proposal, TP 3-85483, George C. Marshall Space Flight Center, NASA, Huntsville, Alabama, May 1963.

3. Effects of High Humidity on Dielectric Properties of Casting Resins, H. K. Graves, M. A. Pizzino, Electrical Manufacturing, April 1956.

4. Development of Improved Thermal Shock Resistant Dielectric Materials for Embedding Electronic Components, R. B. Feuchtbaum, C. J. Bahum, J. B. Rust, Final Report, Contract NObs 84027, June 1961.

5. Physical and Electrical Properties of Epoxy Resins as a Function of Chemical Composition, R. B. Feuchtbaum, Electrochemical Society Meeting, Indianapolis, Indiana, May 1961.

6. A Four Rod Embedded Electrode Test, R. B. Feuchtbaum, Fifth Conference on Electrical Insulations, Chicago, Illinois, September 1963.

7. Development of Improved Heat Sterilizable Potting Compounds, R. B. Feuchtbaum, Second Quarterly Report, Hughes Aircraft Company, Report P63-93, Culver City, California, page 5, 15 January 1964.

8. Infrared Spectra of Plastics and Resins, R. E. Kagarise and L. A. Weinberger, Naval Research Laboratory, Washington, D. C., NRL Report 4369, 26 May 1954.

9. Kunststoffe, 52, 61-6, A. Rost. (1962).

APPENDIX B

PHOTOGRAPH REFERENCE

Figure	Title	Hughes Reference No.
Figure 1.	Four rod, embedded electrode test chamber. A - specimen chamber; B - U-shaped liquid nitrogen cryotrap; C - sealed tip used to exhaust gaseous samples for spectroscopic analysis after test; D - to exhaust manifold.	R-97347
Figure 2.	Construction of heating ovens. Resistance wire is wrapped around a pyrex thimble to form an oven for the thermal exposure of the preliminary evaluation tests. Left to right: thimble and asbestos paper, thimble wrapped with asbestos paper (note circled hook-shaped anchors for the nicrome wire), thimble wound with nichrome wire, completed oven.	R-97340
Figure 3.	Fabrication of uranium glass-tungsten graded glass seals. Left to right: tungsten pins, tungsten pins with vacuum drawn uranium glass sheaths, uranium glass tube, tungsten conducter pins for embedded electrode and thermocouples sealed in test chamber cap.	R-97341
Figure 4.	Uranium glass-tungsten electrode graded glass seals. The glass sheathed pins and the test chamber cap are heated in the glass blower's lathe, and then hot press formed into one piece.	R-97345
Figure 5.	Completed uranium glass-tungsten electrode graded seal. Note how the pins are held in place before fusion by the graphite die.	R-97346

PROGRAM PLAN

RCN-PP-85

PAGE 1 OF 3
PUBL. DATE 28 MAY 1964

PROGRAM TITLE: IMPROVED HEAT-STERILIZABLE POTTING COMPOUND

CUSTOMER: NASA HAC REF. # A-1893

PROGRAM DESCRIPTION: RESEARCH & DEVELOPMENT PROGRAM TO ESTABLISH AN IMPROVED HEAT-STERILIZABLE POTTING COMPOUND

CONTRACT: NAS 8-5499 (CPFF)

PROJECT ENGINEER: R. B. FEUCHTBAUM (27-48)

RELEASING ORGANIZATION: R&DD

SOURCE CODE: 27 61 00

PRODUCT LINE CODE: CXP

PROGRAM NO.: 4721 A A1893

CLASS OF SUB.	PROG. ITEM NO.	CONTRACT NUMBER CONTRACT ITEM NO.	SCHEDULE SECTION	ORG.	HISTORY
1			CONTRACTUAL PLAN		
	1		REQUEST FOR QUOTE RECEIVED	27-61	
	2		897 ISSUED	"	
	3		PROPOSAL SUBMITTED (CPFF)	"	
	4		CONTRACT NEGOTIATED	"	
	5	NAS 8-5499	CONTRACT RECEIVED 1326-375-XXXX	"	
	6		CUSTOMER NOTIFICATION LTR. 85%	"	
1			ENGINEERING PLAN		
	7	1A	COMPREHENSIVE LITERATURE SURVEY	27-48	
	8	1B	POLYMERIC MATERIALS SHALL BE CHOSEN FOR FURTHER TESTS	"	
	9	1C	CANDIDATE POLYMERS SHALL BE MOD. BY ADDITION OF SUITABLE FILLERS IF NECESSARY, ETC.	"	
	10	1D	POLYMERIC MATERIALS SHALL BE SCREENED TO OBTAIN CANDIDATE MATERIALS FOR FURTHER EVALUATION AND MODIFICATIONS.	"	
	11	1E	MOST PROMISING MATERIALS SHALL BE CHARACTERIZED & EVALUATED FOR PROPERTIES STATED IN RFP	"	
	12	1F	PROPOSED ADDITIONAL TESTS PERFORMED	"	
	13	2	REPORTS	"	
	14		MONTHLY LETTER	"	
	15		QUARTERLY PROGRESS	"	
	16		FINAL SUMMARY (DRAFT, FINAL)	"	
	17	3	SAMPLES	"	
	18	3A	DEVELOPED MATERIALS UNDER PROGRAM	"	

Timeline: 1963 J F M A M J J A S O N D; 1964 J F M A M J J A S O N D; 1965 J F M A M J J A S O N D

FORM 9062-CS

CUT OFF DATE: 8 MAY 1964
Revises Issue Dated: 9 MARCH 1964

* A - MASTER PROGRAM PLAN B - PROGRAM PLAN C THRU R - SUB PROGRAM PLAN S - PROGRAM PLAN SUMMARY

G. L. JOHNSON EXT. 3673
R&DD MASTER PROGRAMMER

PROGRAM PLAN

PROGRAM TITLE	PROGRAM DESCRIPTION	PROJECT ENGINEER	PUBL. DATE
IMPROVED HEAT-STERILIZABLE POTTING COMPOUND	RESEARCH & DEVELOPMENT PROGRAM TO ESTABLISH AN IMPROVED HEAT-STERILIZABLE POTTING COMPOUND	R. B. FEUCHTBAUM (27-48)	28 MAY 1964

RELEASING ORGANIZATION	SOURCE CODE			PRODUCT LINE CODE	PROGRAM NO.		
R&DD	27	61	00	CXP	4721	A	A1893

SCHEDULE SECTION

CLASS OF BUS.	PROG. ITEM NO.	CONTRACT NUMBER CONTRACT ITEM NO.	SCHEDULE SECTION	ORG.	HISTORY
1	19		REFER TO PAGE 1		

MANPOWER SECTION

1963

CLASS OF BUS.	PROG. ITEM NO.	CONTRACT NO.	MANPOWER SECTION		ORG.	J	F	M	A	M	J	J	A	S	O	N	D
1	20		ITEMS 7 THRU 19	PLANNED	27-4X							1.0	1.8	2.0	2.1	2.1	2.1
				ACTUAL	"												
				ACTUAL	27-01												
				ACTUAL	20-XX												
4			SALES FORECAST														
(MAN/MONTHS)			DIRECT	PROGRAMMED													
				ACTUAL													
			INDIRECT	PROGRAMMED													
				ACTUAL													
			TOTAL	PROGRAMMED								1.0	1.8	2.0	2.1	2.1	2.1
				ACTUAL								0.4	0.6	1.1	1.8	1.4	1.3

1964

MANPOWER SECTION		ORG.	J	F	M	A	M	J	J	A	S	O	N	D
ITEMS 7 THRU 19	PLANNED	27-4X	2.0	1.9	1.9	1.6	0.8	0.5	0.3					
	ACTUAL	"	1.5	1.6	2.3	2.7								
	ACTUAL	27-01	-	0.3	T	-0-								
	ACTUAL	20-XX	0.2	-	T	-0-								
SALES FORECAST										1.0	1.0	1.0	1.0	1.0
DIRECT	PROGRAMMED													
	ACTUAL													
INDIRECT	PROGRAMMED													
	ACTUAL													
TOTAL	PROGRAMMED		2.0	1.9	1.9	1.6	0.8	0.5	0.3	1.0	1.0	1.0	1.0	1.0
	ACTUAL		1.7	1.9	2.3	2.7								

1965

MANPOWER SECTION		J	F	M	A	M	J	J	A	S	O	N	D
SALES FORECAST		1.0	1.0	1.0	1.0	1.0	1.0	1.0	1.0	1.0	1.0	1.0	1.0
TOTAL	PROGRAMMED	1.0	1.0	1.0	1.0	1.0	1.0	1.0	1.0	1.0	1.0	1.0	1.0

FACILITIES SECT.

CLASS OF BUS.	PROG. ITEM NO.	CONTRACT NO.	FACILITIES SECT.	SQ. FT.	CODES BLDG	CODES M&E	ORG.	PER REQ TOTAL COST/VAL
1	21		NO ADDITIONAL FACILITIES REQUIRED					

FORM 8080 CS

CUT OFF DATE 8 MAY 1964

MASTER PROGRAM PLAN B – PROGRAM PLAN C THRU R – SUB PROGRAM PLAN S – PROGRAM PLAN SUMMARY

PROGRAM PLAN

PROGRAM TITLE: IMPROVED HEAT-STERILIZABLE POTTING COMPOUND

PRODUCT LINE CODE: CYP | PROGRAM NO.: 4721 A

	CLASS OF BUS	PROG ITEM NO	CONTRACT NUMBER CONTRACT ITEM NO.	MATERIEL SECTION	QTY.	CONTROL TYPE	CODES	1963	1964	1965
22	1	22		MATERIALS				J ←	1.0 → M-J	
23		23		TRAVEL (1 RT LA TO SW)				J ←	0.4 → M-J	
24										
25									CUM EXP THRU 4-26-64	
26	GENERAL PURCHASING			PROGRAMMED COMMITMENTS				J ←	1.4 → M-J	
				EXPENDITURES					3.4 K	
27	OUTSIDE PRODUCTION			PROGRAMMED COMMITMENTS						
				PROGRAMMED COMMITMENT EXPENDITURES						
				EXPERIENCED COMMITMENTS						
				EXPERIENCED COMMITMENT EXPENDITURES						

FINANCIAL SECTION

28

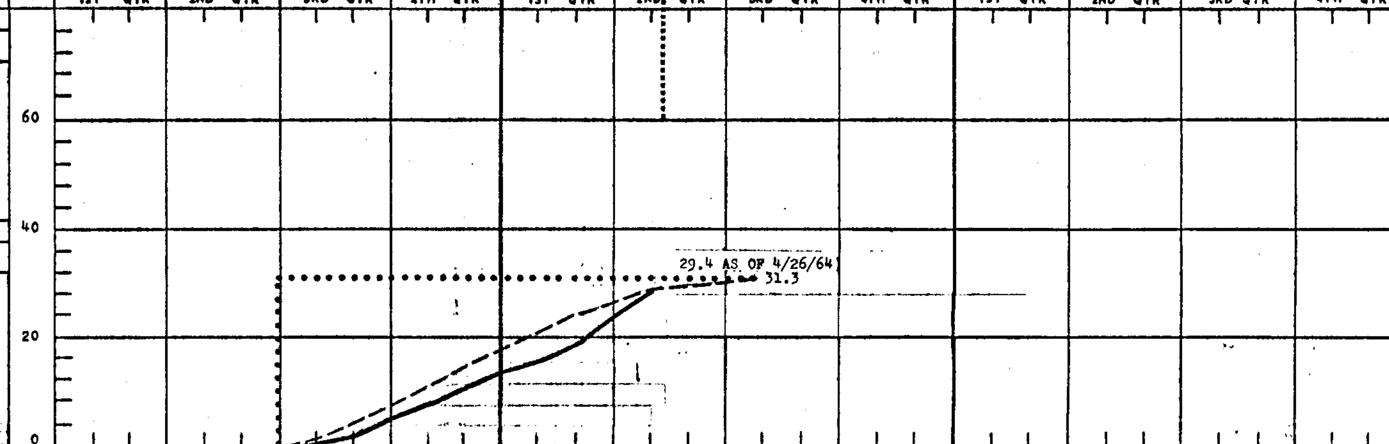
...... AUTHORIZED FUNDS C/A 1326-375-XXXX

SOURCE	DATE AUTHORIZED	AMOUNT AUTHORIZED	CUM. AMOUNT AUTHORIZED
NAS 8-5499 TO 27-4X	6-30-63	31.3	31.3
IDWA #456	(7-12-63)		

ooooo SCHEDULED FUNDS

SOURCE	DATE SCHEDULED	AMOUNT SCHEDULED	CUM. AMOUNT SCHEDULED

AUTHORIZED FUNDS PLUS SCHEDULED FUNDS TOTAL 31.3



	1963 J	A	S	O	N	D	1964 J	F	M	A	M	J	J
— — — PROGRAMMED EXPEND. & COMMITMENTS	1.6	2.8	3.1	3.3	3.3	3.3	3.1	2.9	2.9	2.4	1.3	0.8	0.5
——— ACTUAL EXPEND & COMMITMENTS ⋮ XXX REPROGRAMMED EXPEND. & COMMITMENTS	0.7	1.5	2.1	2.9	3.2	2.2	2.6	3.0	5.7	5.5			

COMMENTS SECTION

29

* A - MASTER PROGRAM PLAN B - PROGRAM PLAN C THRU R - SUB PROGRAM PLAN S - PROGRAM PLAN SUMMARY